Characteristics of First-Order Vortex Lattice Melting: Jumps in Entropy and Magnetization

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We derive expressions for the jumps in entropy and magnetization characterizing the first-order melting transition of a flux line lattice. In our analysis we account for the temperature dependence of the Landau parameters and make use of the proper shape of the melting line as determined by the relative importance of electromagnetic and Josephson interactions. The results agree well with experiments on anisotropic $Y_1Ba_2Cu_3O_{7-\delta}$ and layered $Bi_2Sr_2Ca_1Cu_2O_8$ materials and reaffirm the validity of the London model.

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A cornerstone of the phenomenology of type II superconductors is the Abrikosov mean-field $H-T$ phase diagram comprising a Meissner-Ochsenfeld phase at low fields, a Shubnikov- or mixed phase at intermediate fields, and a normal metallic phase at high fields $H > H_{c_2}$ [1]. Thermal fluctuations may modify this mean-field picture considerably as the vortex lattice melts into a flux-liquid phase, which is the case in high temperature superconductors [2,3]. According to standard symmetry considerations [4], this melting transition is expected to be of first order. Recently, this expectation has been confirmed through the experimental observation of a jump in the magnetization in layered $Bi_2Sr_2Ca_1Cu_2O_8$ (BiSCCO) [5,6] and in anisotropic $Y_1Ba_2Cu_3O_{7-\delta}$ (YBCO) [7] crystals. The latent heat released in the transition has been determined in calorimetric measurements on an YBCO crystal [8,9] and the thermodynamic consistency between the magnetic and calorimetric experiments via the Clapeyron relation has been demonstrated [8].

Though consistency could be achieved in the experiments, simple estimates [6] for the jumps in magnetization and entropy have failed to explain the magnitude and the temperature dependence of these characteristic quantities. In particular, in the layered BiSCCO material the entropy jump per vortex per layer as extracted from the magnetization data via the Clapeyron equation seems to diverge upon approaching the superconducting transition temperature [6], and no explanation of this striking result has been given so far. In this letter we resolve this puzzle and derive the temperature dependence of the jumps in magnetization and entropy. Our results are consistent with all the data measured so far in YBCO and BiSCCO single crystals, see Fig. 1.

Besides construction of simple estimates, the theoretical discussion of the jumps in magnetization and entropy has concentrated on numerical simulations. The problem of the notoriously small entropy jumps obtained in early simulations [10,11] has been cured by the insight of Hu and MacDonald [12], who pointed out the relevance of the temperature dependence of the Ginzburg-Landau parameters in the analysis of numerical data obtained in simulations based on the lowest Landau level approximation. Their insight can be given a much wider perspective, both in terms of its application to other numerical approaches [13] as well as analytic treatment of the problem: combining this concept with our knowledge of the shape of the melting line, we show below how to construct a consistent scheme which provides us with expressions for the jumps in entropy and magnetization, exhibiting both the correct magnitude and temperature dependence. Our analysis then removes recent doubts [12,14] on the ability of a simple London model to describe the large entropy jumps measured on YBCO and BiSCCO crystals close to T_c.

Two important elements in our analysis are the shape of the melting line $H_{\rm m}(T)$ and the relevant fluctuation mode in the vortex system at the melting transition, the latter defining the volume $V_{\rm edf}$ of the elementary degree of freedom. Both quantities depend sensitively on the degree of anisotropy/layeredness of the material through the relative importance of the two types of interactions appearing in the vortex system, the Josephson- and the electromagnetic interaction [15]. As a consequence, the melting process of the vortex lattice in YBCO and BiSCCO exhibits quite distinct characteristics, both regarding the shape of the melting line and the temperature dependence of the jumps in magnetization and entropy. In addition, we will make use of the Clausius-Clapeyron equation relating the jumps in the entropy density Δs and in the magnetic induction ΔB,

$$\Delta s = -\frac{1}{4\pi}\frac{dH_{\rm m}}{dT}\Delta B. \qquad (1)$$

In the following, we first present the analysis for an anisotropic material such as YBCO, where we make use of the scaling form of the London free energy functional which contains the flux lattice constant as the only length scale, rendering the calculation essentially exact.

For strongly layered materials (e.g., BiSCCO), two new length scales, the layer separation d and the London penetration depth λ, become relevant. We show how to construct estimates for the jumps which reduce to the previous results in the scaling regime and which allow us to deal with layered materials.

Consider the statistical mechanics of a system described by an effective free energy functional $\mathcal{F}(T,\phi)$. The coarse grained effective field ϕ (e.g. a Ginzburg–Landau wavefunction) is obtained after integration over the microscopic degrees of freedom, which generates the temperature dependence in $\mathcal{F}$ (e.g. the Landau parameter $\alpha(T)=\alpha(1-T/T_c)$). After integration over the remaining degrees of freedom ϕ we arrive at the partition function (k_B is the Boltzmann constant)

$$Z=\int \mathcal{D}\phi\, e^{-\mathcal{F}(T,\phi)/k_B T}. \tag{2}$$

Taking the derivative of the free energy $F=-k_B T\ln Z$ with respect to T we obtain the entropy,

$$S=S_\circ-\langle\partial_T\mathcal{F}\rangle, \tag{3}$$

where $S_\circ=(\langle\mathcal{F}\rangle-F)/T$ is the configurational entropy of the coarse grained ϕ-field, whereas the second term accounts for the internal temperature dependence in the free energy functional $\mathcal{F}(T,\phi)$. To be specific, let us consider a vortex system within the London approximation where the free energy functional takes the form

$$\mathcal{F}[\{\mathbf{s}_\mu\}]=\varepsilon\varepsilon_\circ a_\circ\sum_{\mu,\nu}\int d\mathbf{s}_\mu\cdot d\mathbf{s}_\nu\frac{e^{-(a_\circ/\lambda)|\mathbf{s}_\mu-\mathbf{s}_\nu|}}{|\mathbf{s}_\mu-\mathbf{s}_\nu|}, \tag{4}$$

with $\varepsilon_\circ=(\Phi_\circ/4\pi\lambda)^2$ the basic energy scale in the problem (proportional to the vortex line energy), λ the London penetration depth, $\varepsilon^2=m/M<1$ is the anisotropy parameter, and $\Phi_\circ=hc/2e$ denotes the flux quantum. In (4) all lengths are measured in units of the vortex separation $a_\circ=\sqrt{\Phi_\circ/B}$, with the configurations expressed through the dimensionless position variables $\mathbf{s}_\mu$. For the vortex system, the parameters $\varepsilon_\circ$ and λ depend on the temperature via $\lambda^2(T)=\lambda_0^2/[1-(T/T_c)^2]$. In the limit $\lambda>a_\circ$ the functional (4) assumes a simple scaling form, with all physical parameters appearing in the prefactor $\varepsilon\varepsilon_\circ(T)a_\circ$, the remaining factor representing a scale independent summation over geometrical configurations of lines [16]. Within the scaling regime, all physical results depend on the combination $\varepsilon\varepsilon_\circ(T)a_\circ$. E.g., the shape $B_\mathrm{m}(T)$ of the melting line is given by the condition $\varepsilon\varepsilon_\circ(T_\mathrm{m})a_\circ/k_B T_\mathrm{m}=$ const. ($=1/2\sqrt{\pi}c_\mathrm{L}^2$, we make use of the usual definition of the Lindemann number c_L), from which one easily derives the standard result [3,15]

$$B_\mathrm{m}(T)\approx\frac{\Phi_\circ}{\lambda^2}4\pi c_\mathrm{L}^4\frac{\varepsilon^2\varepsilon_\circ^2\lambda^2}{(k_B T)^2}\propto\left(1-\frac{T^2}{T_c^2}\right)^2 \tag{5}$$

(note that Eq. (5) derives from scaling and proves the validity of the Lindemann criterion). Returning to (3), we can again make use of the scaling form of $\mathcal{F}$ and express the second term in (3) through the energy $\langle\mathcal{F}\rangle$: $\langle\partial_T\mathcal{F}\rangle=(\partial_T\ln\varepsilon_\circ)\langle\mathcal{F}\rangle$ and inserting back into (3) we arrive at the relation

$$S=S_\circ\left(1-\frac{T}{\varepsilon_\circ}\frac{d\varepsilon_\circ}{dT}\right)+\frac{T}{\varepsilon_\circ}\frac{d\varepsilon_\circ}{dT}\frac{F}{T}. \tag{6}$$

At the phase transition the entropy exhibits a jump while the free energy F remains continuous. From (6) we thus infer the following relation between the entropy jump ΔS and its configurational part $\Delta S_\circ$,

$$\frac{\Delta S}{\Delta S_\circ}=\left(1-\frac{T}{\varepsilon_\circ}\frac{d\varepsilon_\circ}{dT}\right)=\frac{1+(T_\mathrm{m}/T_c)^2}{1-(T_\mathrm{m}/T_c)^2}, \tag{7}$$

where we have made use of the specific temperature dependence of the line energy $\varepsilon_\circ\propto[1-(T/T_c)^2]$ in the last equation. We find, that close to the thermodynamic transition the entropy jump ΔS is strongly enhanced with respect to its configurational component $\Delta S_\circ$. Note that it is the latter quantity which is amenable to simple estimates [6] and which is usually calculated in numerical simulations [13]. The result (7) can be easily understood as the simple consequence of a "coordinate" transformation: the temperature enters the partition function Z not merely as a scale parameter but rather in a combination $T/[1-(T/T_c)^2]$. Close to T_c this expression becomes singular, resulting in a strong enhancement of the entropy jump [17]. The physical origins of this enhanced entropy are microscopic fluctuations: Within the coarse grained vortex model these fluctuations surface in the temperature dependence of the phenomenological parameters in $\mathcal{F}$, which are singular at the mean–field transition.

Next, let us find an expression for the configurational part $\Delta S_\circ$ of the entropy jump: within the scaling regime, standard arguments *dictate* the form

$$\Delta S_\circ=\eta k_B\frac{V}{V_\mathrm{edf}}, \tag{8}$$

with $V_\mathrm{edf}=\varepsilon a_\circ^3$, V denotes the system volume, and η is a small number. Physically, this result can be understood by attributing the thermal energy $k_B T$ to each individual degree of freedom. The volume V_edf is determined by the dominant modes leading to melting, which are located at the Brillouin zone boundary with $k_\perp\approx\sqrt{4\pi}/a_\circ$. Hence, we can define the volume per degree of freedom in the form $V_\mathrm{edf}=a_\circ^2 L$. Furthermore, in an anisotropic material the important fluctuations involve the wavevector $k_z\sim 1/\varepsilon a_\circ$ along the field, thus $L=\varepsilon a_\circ$ (with all numericals absorbed in η). Combining (7) and (8) we obtain the final result for the entropy jump per vortex per layer,

$$\Delta S_d\approx 2\eta\frac{d}{\varepsilon a_\circ}\frac{k_B}{1-(T_\mathrm{m}/T_c)^2}. \tag{9}$$

On the melting line, the product $a_\circ[1-(T_{\rm m}/T_c)^2]$ is (roughly) temperature independent and we find a *constant* but material dependent entropy jump per vortex per layer, in agreement with experiments on YBCO [8].

In order to find a numerical result for the entropy jump we compare the latent heat per vortex line $L_l = T_{\rm m}\Delta S_\circ a_\circ^2$ with the result of numerical simulations carried out within the present London formalism [13]: Making use of (5) and the numerical result $L_l = 0.015\,\varepsilon_\circ$ we find the expression $\Delta S_d \approx 0.03\,\varepsilon_\circ(0)d/T_c$ and using parameters for YBCO ($\lambda \approx 1400$ Å and $d = 12$ Å) we obtain the value $\Delta S_d \approx 0.4\,k_B$, in good agreement with experiment [8]. Using the result for the melting line in Ref. [13], $\varepsilon\varepsilon_\circ(T_{\rm m})a_\circ/k_B T_{\rm m} \approx 11$, we obtain a value for the parameter η, $\eta \approx 0.16$.

Let us turn next to the jump ΔB in the induction. In order to make use of the Clapeyron equation (1) we have to determine the slope $\partial_T B_{\rm m}$ of the melting line [18]. Ignoring for the moment the temperature dependence in the free energy (4) (and thus in the parameters of (5)) we have $\partial_T B_{\rm m} = -2B_{\rm m}/T_{\rm m}$. The full result which accounts for the temperature dependence in $\varepsilon_\circ$ reads

$$\frac{dB_{\rm m}}{dT} = -\frac{2B_{\rm m}}{T_{\rm m}}\left(1-\frac{T}{\varepsilon_\circ}\frac{d\varepsilon_\circ}{dT}\right). \tag{10}$$

The same factor relating the two slopes appears in equation (7) which expresses the entropy jump ΔS through $\Delta S_\circ$. Thus in the final expression for the jump ΔB in the induction this correction factor drops out and we arrive at the simple result (we use $V_{\rm edf} = a_\circ^2 L$)

$$\Delta B = \mu\frac{k_B T_{\rm m}}{\Phi_\circ L}, \tag{11}$$

where $\mu = 2\pi\eta \approx 1.0$. Using $L = \varepsilon a_\circ$ as appropriate in an anisotropic material we arrive at the final result for the jump in B,

$$\Delta B \approx \mu\frac{k_B T_{\rm m}}{\Phi_\circ\varepsilon a_\circ} \approx 6.\,10^{-4}\,\frac{\Phi_\circ}{\lambda^2(T_{\rm m})}. \tag{12}$$

Note that in an incompressible (uncharged, $e \to 0$ and $\lambda \to \infty$) system, we correctly find $\Delta B \to 0$. Rewriting (12) in the form $\Delta B[{\rm G}] \approx (1.5\cdot 10^{-6}/\varepsilon)T_{\rm m}[{\rm K}](B_{\rm m}[{\rm G}])^{1/2}$ and choosing $\varepsilon = 1/8$ we arrive at a good agreement with the magnetization data of Schilling *et al.* [8] on an YBCO single crystal, see Fig. 1 (we have made use of the experimentally measured melting line $B_{\rm m}(T)$).

So far our analysis has been essentially exact: we have exploited the scaling behavior of the London functional in the regime $a_\circ < \lambda$ and have deduced the single unknown parameter η from a comparison to a numerical simulation of the London model. While this approach is successfully applied to a continuous anisotropic superconductor such as YBCO, we have to reconsider the situation for strongly layered materials (e.g., BiSCCO) as new length scales (d, λ) enter the problem. We then can make use of an important insight provided by the above derivation, namely that the calculation of the jump in B does not suffer from the complications associated with the determination of the jump in entropy. This is because the entropy involves a derivative of the free energy with respect to temperature, whereas the induction is given by the derivative with respect to the magnetic field. The latter usually does not show up in the Landau parameters [19]. Indeed, we can arrive at the result (11) starting from the general thermodynamic relation $B = -(4\pi/V)\partial_H G|_T$, where G is the Legendre transform of the free energy F, $G(T,H) = F(T,B) - BHV/4\pi$. With the estimate $G \sim k_B TV/V_{\rm edf}$ and making use of the power law dependence of $V_{\rm edf}$ on H we obtain

$$\Delta B \approx \frac{\mu' k_B T_{\rm m}}{HV_{\rm edf}}. \tag{13}$$

Inserting the ansatz $V_{\rm edf} = a_\circ^2 L = \Phi_\circ L/H$ we immediately recover the result (11) (with μ replaced by an unknown numerical μ'). Once the jump in the induction ΔB is known, we can make use of the Clausius-Clapeyron equation (1) and arrive at the result for the jump in the entropy. The problematic factor arising from the temperature derivative of the free energy functional $\mathcal{F}$, see (3) and (7), is taken care of by the derivative $\partial_T B_{\rm m}$ of the melting line, see (10).

We proceed with the analysis of the jumps in magnetization and entropy for strongly layered superconductors, following the above line of thought. To do so we need to know the length L of the relevant modes at melting, see (11), as well as the shape of the melting curve in a layered superconductor, to be used in (1).

In layered BiSCCO the melting line is pushed down to low fields $B_{\rm m}(T) < B_\lambda(T) = \Phi_\circ/\lambda^2$ over a large portion of the phase diagram. The dominant interaction in the vortex system is then given by the electromagnetic one. The loosely bound pancake vortices undergo large thermal fluctuations and dominate the melting process, hence $L = d$. The shape of the melting line follows most easily from a Lindemann analysis with $\langle u^2\rangle = c_{\rm L}^2 a_\circ^2 \sim Td/\varepsilon_l(k_z \sim 1/d)$, while making use of the dispersive electromagnetic line tension $\varepsilon_l \sim \varepsilon_\circ/\lambda^2 k_z^2$ [15],

$$B_{\rm m}^{\rm em}(T) \approx \frac{\Phi_\circ}{\lambda^2}\frac{c_{\rm L}^2}{2}\frac{\varepsilon_\circ d}{k_B T} \propto \left(1-\frac{T^2}{T_c^2}\right)^2. \tag{14}$$

Close to T_c the Josephson interaction becomes relevant as soon as $\varepsilon\lambda(T) > d$: For $T > T^{\rm em} \approx T_c[1-(\varepsilon\lambda_0/d)^2]^{1/2}$ the dominant fluctuations at melting are cut off on the larger scale $L \sim \varepsilon\lambda$. The Lindemann criterion takes the form $\langle u^2\rangle = c_{\rm L}^2 a_\circ^2 \sim T\varepsilon\lambda/\varepsilon_l(k_z \sim 1/\varepsilon\lambda)$ and we obtain a melting line following a $(1-T^2/T_c^2)^{3/2}$ behavior [15],

$$B_{\rm m}^{\rm em,J}(T) \approx \frac{\Phi_\circ}{\lambda^2}\frac{\pi c_{\rm L}^2}{4}\frac{\varepsilon\varepsilon_\circ\lambda}{k_B T} \propto \left(1-\frac{T^2}{T_c^2}\right)^{3/2}. \tag{15}$$

Inserting the above results for L into (11) we obtain the jump ΔB in the induction for a *layered* material,

$$\Delta B \approx \begin{cases} \mu' \dfrac{k_B T_{\rm m}}{\Phi_\circ d}, & T_{\rm m} < T^{\rm em}, \\ \mu' \dfrac{k_B T_{\rm m}}{\Phi_\circ \varepsilon \lambda_0} \sqrt{1-(T_{\rm m}/T_c)^2}, & T^{\rm em} < T_{\rm m}. \end{cases} \quad (16)$$

Note that it is the temperature dependence of L, which goes from $\varepsilon a_\circ(T_{\rm m})$ in a continuous anisotropic superconductor to $L = d$ and $L = \varepsilon\lambda(T_{\rm m})$ in a layered material, that leads to the different dependencies in the jump $\Delta B(T)$.

The result (16) explains the experimental observations of Zeldov *et al.* [6], see Fig. 1: At low temperatures $T_{\rm m} < T_c - 7$ K the jump ΔB increases linearly with temperature. About 7 K before reaching the transition, ΔB drops sharply and vanishes at T_c. This behavior is explained in terms of the crossover at $T^{\rm em}$, where the Josephson coupling between the layers becomes relevant and cuts off the further growth of fluctuations. With the same value $\mu' = \mu$ (≈ 1) as above, we rewrite (16) in the form $\Delta B[{\rm G}] \approx (0.07/d[\text{Å}])T_{\rm m}[{\rm K}]$ and $\Delta B[{\rm G}] \approx (0.07/\varepsilon\lambda_0[\text{Å}])T_{\rm m}[{\rm K}][1-(T_{\rm m}/T_c)^2]^{1/2}$ and using the parameters $d = 15$ Å, $\lambda_0 \approx 2000$ Å, and $\varepsilon \approx 1/400$ we obtain excellent agreement with the experimental result of Zeldov *et al.* [6]. Note that the experimental finding $\Delta B(T \to T_c) \to 0$ (while B remains $\gg \Delta B$), combined with (11), hints at a divergence $L(T \to T_c) \to \infty$ and thus a 3D transition as T_c is approached. Finally, the expressions for the jumps in entropy follow from the Clapeyron equation (using the line shapes (14) and (15)),

$$\Delta S_d \approx \begin{cases} \dfrac{\mu}{\pi} \dfrac{k_B}{1-(T_{\rm m}/T_c)^2}, & T_{\rm m} < T^{\rm em}, \\ \dfrac{3\mu}{4\pi} \dfrac{d}{\varepsilon\lambda_0} \dfrac{k_B}{\sqrt{1-(T_{\rm m}/T_c)^2}}, & T^{\rm em} < T_{\rm m}. \end{cases} \quad (17)$$

Unlike in the anisotropic case, for the layered material the entropy jump per vortex per layer diverges on approaching the transition at T_c, again in agreement with the experimental observation [6] (note that at low temperatures the entropy jump in Ref. [6] vanishes as the melting line flattens, possibly due to disorder).

In summary, our new estimates (9), (12) and (16), (17) provide a consistent explanation of the observed characteristic jumps at the first-order melting transition of the vortex crystal in type II superconductors.

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[17] It follows that this analysis is not particular to the lowest Landau level approximation as claimed in Ref. [12].
[18] We ignore the small difference between B and H.
[19] Within the London description of type II superconductors the dependence on $1-(T/T_c)^2 - H/H_{c_2}(0)$ modifies the melting line. However, since $H_{\rm m}/H_{c_2}$ remains small, our simplified discussion is quantitatively correct.

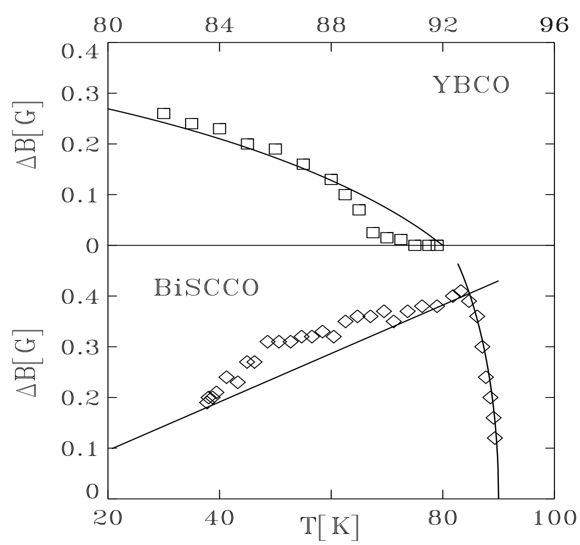

FIG. 1. Top: the jump ΔB in the induction versus temperature T as measured in an YBCO single crystal [8] and calculated from the expression (12). The deviations close to the transition are possibly due to sample inhomogeneity [7]. Bottom: the same for a BiSCCO single crystal [6] using the result (16). The drop in ΔB on approaching T_c is explained in terms of a temperature dependent cutoff in the electromagnetic fluctuations through the Josephson coupling at temperatures $T > T^{\rm em}$. The entropy jump ΔS_d per vortex per layer as obtained from ΔB through the Clapeyron equation diverges at the transition [6], in agreement with the result (17).